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AUTHOR(S):

Miyazaki, Kunimasa

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BROWNIAN MOTION OF A SPHERE IN SLOW SHEAR FLOW

Kunimasa Miyazaki

Dept. of Physical Chemistry, National Institute of Materials and Chemical Research, MITI,
Tsukuba, Ibaraki 305, Japan

The theory of Brownian motion of a particle in a fluid at rest is one of the most brilliant success of the statistical mechanics around the equilibrium state. While the theory around the equilibrium state has been well-established and is impeccable, extension of the theory to the system far from equilibrium seems to remain at primitive level and to be rather rare. It is, in part, due to its difficulty of analysis. Our purpose here is to give a theory of Brownian motion in a nonequilibrium steady states equilibrium starting with as rigorous basis as possible.

We consider the spherical, rigid, and electro-magnetically neutral particle immersed in an incompressible fluid subject to a shear flow given by

$$\mathbf{v}(\mathbf{r}) = \boldsymbol{\beta} \cdot \mathbf{r}, \quad (1)$$

where $\boldsymbol{\beta}$ is the constant traceless tensor. We start with constructing the Langevin equation which describes the stochastic behavior of the sphere.

First we evaluate the friction force exerted on the sphere. In the absence of the shear flow, the force is given by well-known Stokes law, $\mathbf{F}(\omega) = -\xi(\omega)\mathbf{u}(\omega) = -6\pi a\eta(1 + \sqrt{-i\rho\omega/\eta a})\mathbf{u}(\omega)$ (for small ω), where $\mathbf{F}(\omega)$ is the friction force, $\xi(\omega)$ is the friction coefficient, ρ and η is the density and shear viscosity of ambient fluid, respectively, a is the radius of the sphere, and $\mathbf{u}(\omega)$ is the velocity of the sphere. We extended this result to the case in the presence of shear flow using the induced force method[1]. The friction force is expressed as

$$\mathbf{F}(\omega) = -\boldsymbol{\xi}(\omega) \cdot [\mathbf{u}(\omega) - \boldsymbol{\beta} \cdot \mathbf{R}(\omega)], \quad (2)$$

where \mathbf{R} is the position of the sphere. The friction coefficient $\boldsymbol{\xi}(\omega)$ now becomes a tensor and given by

$$\boldsymbol{\xi}(\omega) = \left[\int d\mathbf{r} \int d\mathbf{r}' \int_0^\infty dt e^{i\omega t} \delta(r-a) \mathbf{G}(\mathbf{r} - \mathbf{r}', t) \delta(r'(t)-a) \right]^{-1}, \quad (3)$$

where $\mathbf{G}(\mathbf{r} - \mathbf{r}', t)$ is the Green function of the (linearized) Navier-Stoke equation and $\mathbf{r}(t) \equiv \exp[\boldsymbol{\beta}t] \cdot \mathbf{r}$. This formula is valid up to the first order in $\sqrt{Re} \equiv \sqrt{\beta\rho a^2/\eta} \ll 1$ and $\sqrt{\omega\rho a^2/\eta}$, where Re is the Reynolds number and $\beta \equiv |\boldsymbol{\beta}|$.

Next, using the above expression for the friction coefficient and the theory of fluctuating hydrodynamics, we derive a Langevin equation;

$$-i\omega m\mathbf{u}(\omega) = -\boldsymbol{\xi}(\omega) \cdot [\mathbf{u}(\omega) - \boldsymbol{\beta} \cdot \mathbf{R}(\omega)] + \mathbf{F}_R(\omega), \quad (4)$$

where $\mathbf{F}_R(\omega)$ is the random force. We show that the fluctuation-dissipation theorem is *not valid* in the presence of the macroscopic shear flow. The autocorrelation function of the random force is given by

$$\langle \mathbf{F}_R(\omega) \mathbf{F}_R^*(\omega') \rangle = k_B T [\boldsymbol{\xi}(\omega) + \delta \boldsymbol{\xi}(\omega) + (\text{H.C.})] 2\pi \delta(\omega - \omega'), \quad (5)$$

where $\delta \boldsymbol{\xi}(\omega)$ is a function which can not express in terms of the friction coefficient alone and (H.C.) denotes the Hermite conjugate. $\delta \boldsymbol{\xi}(\omega)$ is the same order in magnitude as the modification of $\boldsymbol{\xi}(\omega)$ due to the shear flow and, therefore, can not be neglected.

Due to the strong coupling with the convection term which appears in eq.(4), the diffusion constant can not be derived from the mean square displacement. We also derive an alternative expression equivalent to the Langevin equation, i.e., the diffusion equation. The diffusion equation for the probability distribution function, $P(\mathbf{r}, t)$, is given by

$$\left\{ \frac{\partial}{\partial t} + \mathbf{v}(\mathbf{r}) \cdot \nabla \right\} P(\mathbf{r}, t) = \int_{-\infty}^t dt' \nabla(t-t') \cdot \mathbf{D}(t-t') \cdot \nabla(t-t') P(\mathbf{r}(t-t'), t'), \quad (6)$$

where $\nabla(t) \equiv \partial/\partial \mathbf{r}(t)$. In this expression, \mathbf{D} is the diffusion coefficient tensor which depends on both time(frequency) and shear rate and is given in terms of the velocity autocorrelation function as

$$\mathbf{D}(t) = \exp[-\beta t] \cdot \langle \delta \mathbf{u}(t) \delta \mathbf{u}(0) \rangle, \quad (7)$$

where $\delta \mathbf{u}(t) \equiv \mathbf{u}(t) - \beta \cdot \mathbf{R}(t)$. This is the generalized Green-Kubo formula for the diffusion coefficient. Substituting the solution of eq.(4) into this expression one obtains

$$\mathbf{D}(t) = k_B T \exp[-\beta t] \cdot \{ \boldsymbol{\mu}(t) + \delta \boldsymbol{\mu}(t) \}, \quad (8)$$

where $\boldsymbol{\mu}(t) \equiv \boldsymbol{\xi}^{-1}(t)$ is the mobility tensor and $\delta \boldsymbol{\mu}(t) \equiv \boldsymbol{\mu}(t) \cdot \delta \boldsymbol{\xi}(t) \cdot \boldsymbol{\mu}^\dagger(t)$. This is the generalized Einstein-Stokes relation.

The diffusion coefficient features behavior common to transport coefficients for the fluidal system in steady shear flow[2];

$$\mathbf{D}(0, \beta) \sim D_0 - D' \sqrt{\beta}, \quad \text{for small } \omega, \quad \mathbf{D}(\omega, 0) \sim D_0 - D'' \sqrt{\omega}, \quad \text{for small } \beta, \quad (9)$$

where $D_0 \equiv k_B T / 6\pi a \eta$ is the Einstein-Stokes diffusion coefficient.

Asides of the above argument, the relevance of the friction coefficient with the principle of Material Frame Indifference(MFI) was also discussed[3].

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